

## Curved and diffuse interface effects on the nuclear surface tension

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We redefine the surface tension coefficient for a nuclear Fermi-liquid drop with a finite diffuse layer. Following the Gibbs-Tolman concept, we introduce the equimolar radius  $R_e$  of the droplet surface at which the surface tension is applied and the radius of tension surface  $R_s$  which provides the minimum of the surface tension coefficient  $\sigma$ . This procedure allows us to derive both the surface tension and the corresponding curvature correction (Tolman length) correctly for the curved and diffuse interface. We point out that the curvature correction depends significantly on the finite diffuse interface. We show that Tolman's length  $\xi$  is negative for a nuclear Fermi-liquid drop. The value of the Tolman length is only slightly sensitive to the Skyrme force parametrization and equals  $\xi = -0.36$  fm.

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### I. INTRODUCTION

The binding energy of saturated many-particle systems like a nucleus exhibits the hierarchy of series in powers of mass number  $A^{1/3}$  which is associated with well-established experimental features and plays an important role in the understanding of macroscopic properties of nuclei [1]. In a simplest case, the surface energy  $E_S$  of such systems is given by the term of order  $A^{2/3}$  in this hierarchy. The structure of the surface energy  $E_S$  and the corresponding surface tension coefficient  $\sigma$  depend on the interparticle interaction and the surface conditions. Moreover, the nucleus is a two component, charged system with a finite diffuse layer. This fact specifies a number of various peculiarities of the nuclear surface energy  $E_S$ : dependency on the density profile function, contribution to the surface symmetry energy, connection to the nuclear incompressibility, etc. The additional refinements of  $E_S$  appear due to the quantum effects arising from the smallness of a nucleus. In particular, the curved interface creates the curvature correction to  $E_S$  of order  $A^{1/3}$  and can play an appreciable role in small nuclei.

The curvature correction to the planar tension coefficient and the corresponding Tolman length [2] can be estimated phenomenologically using the polynomial, in powers of  $A^{1/3}$ , expansion of mass formula [3–6]. However the influence of the curved interface on the properties of small quantum systems is still poorly studied because of the finite diffuse layer where particle density drops down to the zero value. The presence of the finite diffuse layer in a small drop creates, at least, two questions: (i) What is the actual radius of a drop? (ii) What is the physical surface where the surface tension is applied? Because of the presence of the diffuse layer, different definitions for the size of the drop are possible [7] which all give the value of drop radius located within the diffuse layer. Note also that though the width of diffuse layer is much less than the range of approximate uniformity of the particle density, one still needs the strict definition of the drop size because of the following reason. In contrast to the planar geometry, the area  $S$  for the spherical (curved) surface will depend on the choice of drop radius and this will affect the value of the surface tension  $\sigma$  derived from the surface energy.

Gibbs was the first who addressed the problem of the correct definition of the radius and the surface of tension to a small drop with a diffuse interface [9]. After him, Tolman drew attention [2] to the fact that two different radii have to be introduced in this case: the equimolar radius  $R_e$ , which gives the actual size of the corresponding sharp-surface droplet for a given particle number  $A$ , and the radius of tension  $R_s$ , which derives, in particular, the capillary pressure, see below in Sec. II. Following Tolman, see also Ref. [8], the surface tension  $\sigma_e \equiv \sigma(R_e)$  approaches the planar limit  $\sigma_\infty$  as

$$\sigma(R_e) = \sigma_\infty \left( 1 - \frac{2\xi}{R_e} + \mathcal{O}(R_e^{-2}) \right), \quad (1)$$

where  $\xi$  is the Tolman's length [2]. At the same time the capillary pressure  $P_{\text{capil}}$ , which is generated by the curved surface and provides the equilibrium condition for the well-defined radius  $R_e$ , is determined by the radius of tension  $R_s$  [9] [see also below Eqs. (17) and (18)]

$$P_{\text{capil}} = \frac{2\sigma}{R_s}. \quad (2)$$

In general, the presence of the curved interface affects both the bulk and the surface properties. The curvature correction  $\Delta\sigma_{\text{curv}} = -2\sigma_\infty\xi/R_e \sim A^{-1/3}$  is usually negligible in heavy nuclei. However, this correction can be important in some nuclear processes. For example the yield of fragments at the nuclear multifragmentation or the probability of clusterization of nuclei from the freeze-out volume in heavy ion collisions are derived by the statistical weight  $W$  of the radius fluctuations [10]. In both above mentioned processes, small nuclei necessarily occur and the exponential dependence of the statistical weight  $W$  on the surface tension  $\sigma$  [10] should cause a sensitivity of both processes to the curvature correction  $\Delta\sigma_{\text{curv}}$ .

In nuclear physics, the curvature correction to the surface tension was intensively investigated phenomenologically [3,4,6] as well as within the quantum approaches [11]. Using the Thomas-Fermi approximation (TFA), the dependence on curvature of the nuclear surface energy was studied in Ref. [12] and the various terms of the droplet model were derived from the Skyrme interaction in Ref. [13]. We point out that within the

TFA, the nucleus is considered as a quantum liquid. In contrast to the classical liquid, where the van der Waals equation of state and the corresponding saturation properties are caused by the short-range interparticle correlations (collisions), the nuclear quantum liquid is collisionless. The fundamental saturation property is provided there by the self-consistency conditions and the specific density-dependent effective interaction. Moreover, the nuclear quantum liquid is a liquid in a space of probability distribution, current probability, etc. [14]. This fact allows one to extend the consideration of liquid properties to the case of quite small nuclei of order  $A \sim 10$  until the saturation properties are not violated.

The curvature correction can also play an appreciable role for the nuclear fission near the scission point and for the nuclear fusion in the neck region. This aspect of large nuclear deformations was not studied yet. In Ref. [15], a general procedure restricted by the Skyrme-type functional and the terms of the order  $\hbar^2$  was applied for calculation of the curvature energy and in Ref. [16] the curvature-energy was studied by use of semiclassical mean-field approaches with including higher-order terms. In Ref. [17] a closed expression for the nuclear curvature energy and its expansion into series of volume terms and surface moments was deduced in a soluble model. For the Fermi gas model in an external Woods-Saxon potential the curvature energy was calculated in Ref. [18]. A special care was given to the kinetic energy operator and the bulk density oscillations within the quantum-mechanical approach [11]. It was shown that, in contrast to the semiclassical approaches, to obtain a reasonable value for the curvature energy the particle and energy densities should be averaged in a special way.

In the present paper, we suggest the microscopic analysis of the curvature correction to the surface tension of a small drop with a finite diffuse layer. We follow the ideology of the extended Thomas-Fermi approximation (ETFA) with effective Skyrme-like forces combining the ETFA and the direct variational method. In our consideration, the proton and neutron densities  $\rho_p(\mathbf{r})$  and  $\rho_n(\mathbf{r})$  are generated by the diffuse-layer profile functions which are eliminated by the requirement that the energy of the nucleus should be stationary with respect to variations of these profiles. In order to formulate proper definition for the drop radius, we will use the concept of the dividing surface, originally introduced by Gibbs [9]. Following Gibbs, we will introduce the superficial (surface) density as the difference (per unit area of dividing surface) between actual number of particles  $A$  and the number of particles  $A_V$  which a drop would contain if the particle density were uniform.

This paper is organized as follows. In Sec. II we give the thermodynamical derivation of the surface tension for a finite system. The Tolman length is derived in Sec. III. Numerical results and conclusions are summarized in Secs. IV and VI. The connection of Gibbs-Tolman (GT) approach to the droplet model is given in Sec. V.

## II. EQUIMOLAR SURFACE

We will calculate the dependence of the surface tension coefficient on the position of the dividing surface in a small

Fermi-liquid drop with a finite diffuse layer similarly to the procedure described in Refs. [8,19]. The goal of the calculations is to determine the position of the equimolar surface, the dependence of surface tension on the bulk density and the sensitivity of the curvature correction (Tolman length  $\xi$ ) to the parametrization of the effective nuclear forces.

We consider the uncharged symmetric ( $N = Z$ ) droplet having the number of particles  $A = N + Z$ , the chemical potential  $\lambda$  and the free energy  $F$ . Note that the thermodynamical consideration is most adequate here because of the finite diffuse interface in a cold nucleus is similar to the vapor environment in a classical liquid drop. In order to formulate proper definition for the drop radius, we will use the concept of dividing surface of radius  $R$ , originally introduced by Gibbs [9]. Following Refs. [8,9], we introduce the formal (arbitrary but close to the interface) dividing surface of radius  $R$ , the corresponding volume  $\mathcal{V} = 4\pi R^3/3$  and the surface area  $S = 4\pi R^2$ . The droplet free energy  $F$  will be then split between the volume,  $F_V$ , and surface,  $F_S$ , parts

$$F = F_V + F_S, \quad (3)$$

where

$$F_V = (-P + \lambda \varrho_V) \mathcal{V}, \quad F_S = (\sigma + \lambda \varrho_S) S. \quad (4)$$

Here  $P = P(\lambda)$  is the pressure of nuclear matter achieved at some volume particle density  $\varrho_V = A_V/\mathcal{V}$  and  $\varrho_S = A_S/S$  is the surface particle density, where  $A_V$  and  $A_S$  are the volume and the surface particle numbers, respectively. Irrespective to the way of calculation of the total free energy  $F$ , its volume part  $F_V$  stands for the nuclear matter free energy of the uniform density  $\varrho_V$  within the volume  $\mathcal{V}$ . The state of the nuclear matter inside the specified volume is chosen to have the chemical potential  $\lambda$  equal to that of the actual (in the presence of the finite diffuse layer) drop. Having the value of  $\lambda$  one can calculate all the intensive quantities like the free energy per particle  $F_V/A_V$ , the particle density  $\varrho_V$  and the pressure  $P = -\partial F_V/\partial \mathcal{V}|_{A_V}$  from the equation of state for the infinite nuclear matter. The surface part of the free energy  $F_S$  as well as the surface particle number  $A_S$  are considered as the excess quantities responsible for “edge” effects with respect to the corresponding volume quantities. Thus, the chemical potential  $\lambda$  is the key quantity needed to determine both the volume part of the free energy  $F_V$  and the surface energy  $F_S$  due to Eq. (3).

The actual particle number is given by

$$A = A_V + A_S = \varrho_V \mathcal{V} + \varrho_S S. \quad (5)$$

Note also that the surface (superficial) particle number  $A_S$  is a formal quantity which is caused by the deviation of the volume part  $A_V$  from the actual particle number  $A$  through an arbitrary choice of the dividing surface. The value of  $A_S$  disappears for the actual size of the sharp-surface droplet given by the equimolar radius  $R_e$ , see Ref. [9] and below in Sec. III. The use of Eqs. (3)–(5) gives the following relation for the surface tension:

$$\sigma = \frac{F - \lambda A}{S} + \frac{PV}{S} = \frac{\Omega - \Omega_V}{S}, \quad (6)$$

where symbol  $\Omega = F - \lambda A$  stands for the grand potential and  $\Omega_V = -P_V$ . To reveal a  $R$ -dependence of the surface tension  $\sigma$ , it is convenient to introduce the grand potential per particle  $\omega = F/A - \lambda$  for the actual droplet and  $\omega_V = F_V/A_V - \lambda = -P/\varrho_V$  for the volume part. Then the surface tension is written as

$$\sigma[R] = \frac{\omega A}{4\pi R^2} - \frac{1}{3}\omega_V \varrho_V R. \quad (7)$$

Here, the square brackets denote a dependence of the observable  $F$ ,  $\lambda$ ,  $P$ , etc. on the dividing surface radius  $R$  which is different than the dependence on the physical size of a droplet [19]. Using Eq. (6), the capillary pressure  $P$  reads

$$P = 3 \frac{\sigma[R]}{R} - 3 \frac{F - \lambda A}{4\pi R^3}. \quad (8)$$

Taking the derivative from Eq. (8) with respect to the formal dividing radius  $R$  and using the fact that the observable quantities  $F$ ,  $\lambda$ , and  $P$  should be  $R$  independent (changing dividing radius  $R$  we keep the particle density invariable), one can rewrite Eq. (8) as

$$P = 2 \frac{\sigma[R]}{R} + \frac{\partial}{\partial R} \sigma[R], \quad (9)$$

which is the generalized Laplace equation.

The choice of the dividing radius  $R$  is arbitrary, the only condition is to keep the same chemical potential  $\lambda$ . So, the formal value of surface density  $\varrho_S$  can be positive or negative depending on  $R$ . From Eq. (5) one finds

$$\varrho_S[R] = \frac{A}{4\pi R^2} - \frac{1}{3}\varrho_V R. \quad (10)$$

We have performed the numerical calculations using Skyrme type of the effective nucleon-nucleon interaction. The energy and the chemical potential for actual droplets have been calculated using a direct variational method within the extended Thomas-Fermi approximation [20]. The free energy  $F$  of the nucleus is given by the following functional:

$$F = \int d\mathbf{r} \{ \epsilon_{\text{kin}}[\rho_n, \rho_p] + \epsilon_{\text{Sk}}[\rho_n, \rho_p] + \epsilon_C[\rho_p] \}, \quad (11)$$

where  $\epsilon_{\text{kin}}[\rho_n, \rho_p]$  is the kinetic energy density,  $\epsilon_{\text{Sk}}[\rho_n, \rho_p]$  is the potential energy density of Skyrme  $NN$ -interaction and  $\epsilon_C[\rho_p]$  is the Coulomb energy density. The equilibrium condition can be written as a Lagrange variational problem. Namely,

$$\delta(F - \lambda_n N - \lambda_p Z) = 0, \quad (12)$$

where the variation with respect to all possible small changes of  $\rho_n$  and  $\rho_p$  is assumed.

Assuming the leptodermous condition, the total energy takes the following form of  $A$ ,  $X$  expansion:

$$F/A \equiv e_A = e_0(A) + e_1(A)X + e_2(A)X^2, \quad (13)$$

where  $X$  is the isotopic asymmetry parameter  $X = (N - Z)/A$  and

$$e_i(A) = c_{i,0} + c_{i,1}A^{-1/3} + c_{i,2}A^{-2/3}. \quad (14)$$

An explicit form of coefficients  $c_{i,j}$  for the Skyrme forces is given in Ref. [20]. Using the trial profile function for the

neutron  $\rho_n(r)$  and proton  $\rho_p(r)$  densities and performing the direct variational procedure, we can evaluate the equilibrium particle densities  $\rho_{\pm}(r) = \rho_n(r) \pm \rho_p(r)$ , the equilibrium bulk density  $\rho_{\pm,0} = \lim_{r \rightarrow 0} \rho_{\pm}(r)$ , the total free energy per particle  $F/A$ , and the chemical potentials  $\lambda_n$  and  $\lambda_p$ , see Ref. [20] for details. From now on, we will consider symmetric and uncharged nuclei with  $\lambda_n = \lambda_p = \lambda$  and zero asymmetry parameter  $X = 0$ .

The volume part of free energy  $F_V/A_V$  is associated with the coefficient  $c_{0,0}$  of Ref. [20]

$$\begin{aligned} F_V/A_V &= c_{0,0}, \\ c_{0,0} &= \frac{\hbar^2}{2m} \alpha \rho_{+,0}^{2/3} + \frac{3t_0}{8} \rho_{+,0} + \frac{t_3}{16} \rho_{+,0}^{\nu+1} \\ &\quad + \frac{\alpha}{16} [3t_1 + t_2(5 + 4x_2)] \rho_{+,0}^{5/3}, \end{aligned} \quad (15)$$

where  $\alpha = (3/5)(3\pi^2/2)^{2/3}$  and  $t_i$ ,  $x_2$  and  $\nu$  are the Skyrme force parameters. Using the evaluated chemical potential  $\lambda$ , we fix the particle density  $\varrho_V = \varrho_V(\lambda)$  from the condition

$$\left. \frac{\partial F_V}{\partial A_V} \right|_V = \left. \frac{\partial}{\partial \rho_{+,0}} (\rho_{+,0} c_{0,0}) \right|_{\rho_{+,0} = \varrho_V} = \lambda. \quad (16)$$

For an arbitrary dividing radius  $R$  we evaluate then the volume particle number  $A_V = 4\pi R^3 \varrho_V/3$  and the volume part of free energy  $F_V/A_V$ . Finally, evaluating the surface parts  $A_S = A - A_V$  and  $F_S = F - F_V$ , we obtain the surface tension coefficient  $\sigma[R]$  for an arbitrary radius  $R$  of dividing surface. The dependence of the surface tension  $\sigma[R]$  on the location of the dividing surface for  $A = 208$  is shown in Fig. 1. As seen from Fig. 1, function  $\sigma[R]$  has a minimum at radius  $R = R_S$  (radius of the surface of tension [8]) which usually does not coincide with the equimolar radius  $R_e$ . The radius  $R_S$  denotes the location within the interface. Note that for  $R = R_S$  the capillary pressure of Eq. (9) satisfies the classical Laplace relation

$$P = 2 \left. \frac{\sigma[R]}{R} \right|_{R=R_S}. \quad (17)$$

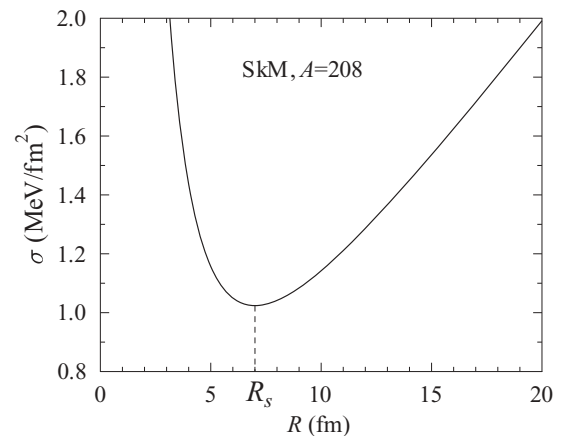


FIG. 1. Surface tension  $\sigma$  as a function of the dividing radius  $R$  for  $A = 208$ . The calculation was performed using the SkM force.  $R_S$  denotes the dividing radius where  $\sigma$  approaches the minimum value.

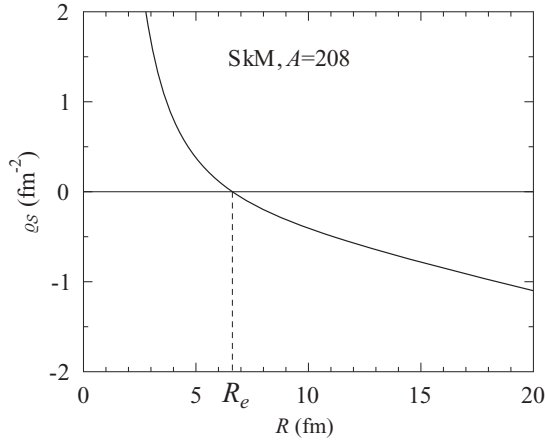


FIG. 2. Surface particle density  $\rho_S$  versus dividing radius  $R$  for  $A = 208$ . The calculation was performed using the SkM force.  $R_e$  denotes the equimolar radius where  $\rho_S$  becomes zero.

### III. SURFACE TENSION AND TOLMAN LENGTH

In Fig. 2 we present the calculation of the surface particle density  $\rho_S[R]$ . Note that, in general, the surface free energy  $F_S$  includes both contributions from the surface tension  $\sigma$  itself and from the bulk binding energy of  $A_S$  particles within the surface layer. The equimolar surface and the actual physical radius  $R_e$  of the droplet are derived by the condition  $\rho_S[R_e] = 0$  [2,8,19], i.e., the contribution from the bulk binding energy should be excluded from the surface free energy  $F_S$ . The corresponding radius is marked in Fig. 2. Equimolar dividing radius  $R_e$  defines the physical size of the sharp surface droplet and the surface at which the surface tension is applied.

Figure 3 illustrates the profile density of the droplet (solid line) and its volume part (dashed line). One can see from this figure that the density of nuclear matter,  $\rho_V$ , slightly differs from the droplet bulk density,  $\rho_{+,0}$ . Using the SkM force, we obtain for  $A = 208$  slightly different values of

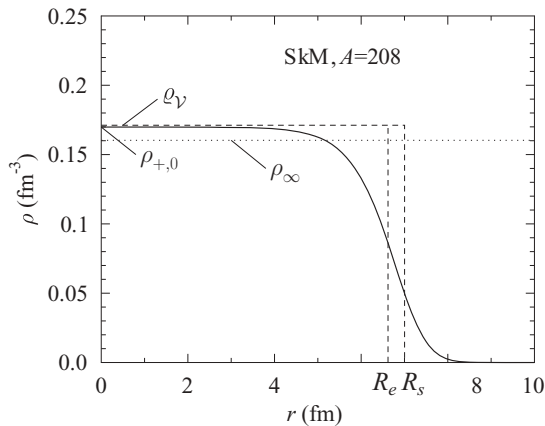


FIG. 3. Profile density  $\rho(r)$  for  $A = 208$ . Solid line shows calculation for the actual droplet, dashed line represents the distributions which correspond to the equimolar and tension dividing surfaces, dotted line is the particle density  $\rho_\infty$  in nuclear matter. The calculation was performed using the SkM force.  $R_e$  denotes the equimolar radius,  $R_s$  is the radius of the surface of tension.

$\rho_V = 0.171 \text{ fm}^{-3}$  and  $\rho_{+,0} = 0.170 \text{ fm}^{-3}$ . This difference disappears for an incompressible liquid or in the planar limit. In Ref. [3] the approximation  $\rho_{+,0} = \rho_V$  was used to obtain the curvature correction to the surface tension. Since the correction for curvature is calculated in the limit of semi-infinite matter, such approximation should, obviously, give correct results. Note also that both particle densities  $\rho_V$  and  $\rho_{+,0}$  exceed the nuclear matter density  $\rho_\infty$  (dotted line in Fig. 3). That is because the surface pressure, which influences the bulk properties, leads to an increase in the nucleon density in center of the nucleus.

Considering an arbitrary choice of the dividing surface and following the Gibbs-Tolman concept, we have determined two radii, the equimolar dividing radius  $R_e$  which corresponds to zero surface density  $\rho_S$  and the radius of tension  $R_s$  which corresponds to the minimum value of the surface tension. From Eqs. (7) and (10) the values of these radii are given by

$$R_e = \left( \frac{4\pi\rho_V}{3A} \right)^{-1/3}, \quad R_s = \left( -\frac{2\pi\rho_V}{3A} \frac{\omega_V}{\omega} \right)^{-1/3}. \quad (18)$$

Following Gibbs and Tolman [2,9], we will assume that the physical (measurable) value of the surface tension is that taken at the equimolar dividing surface. Taking Eq. (9) for  $R = R_s$ , using Eqs. (17) and (1) and introducing small value  $\eta = R_e - R_s$ , we obtain

$$P = \frac{2\sigma_\infty}{R_s} \left( 1 - \frac{2\xi}{R_s} + \mathcal{O}(R_s^{-2}) \right). \quad (19)$$

Taking Eq. (9) for  $R = R_e$  and Eq. (1) we find

$$P = \frac{2\sigma_\infty}{R_s} \left( 1 - \frac{\xi + \eta}{R_s} + \mathcal{O}(R_s^{-2}) \right). \quad (20)$$

We point out, that one should make a difference between the formal derivative  $\sigma'[R]$  in Eq. (9) and the derivative  $\sigma'(R)$  where the surface tension is treated as a function of physical size. However, for the special case of the equimolar dividing surface one can derive  $\sigma'[R_e] = \sigma'(R_e)$ , see [8]. In particular, using Eq. (1) one finds  $\sigma'(R_e) = \sigma_\infty(2\xi R_e^{-2} + \mathcal{O}(R_e^{-3}))$  and, in contrast to  $\sigma'[R_s] = 0$ , one has  $\sigma'(R_s) = \sigma_\infty(2\xi R_s^{-2} + \mathcal{O}(R_s^{-3}))$ . Comparing Eqs. (19) and (20) for  $R_s \rightarrow \infty$ , we obtain the Tolman result [2] (see also [19])

$$\xi = \lim_{A \rightarrow \infty} (R_e - R_s). \quad (21)$$

This result leads to the important conclusions which were not mentioned in previous studies of nuclear surface. First, one needs to define two different radii: the equimolar radius,  $R_e$ , for a proper extraction of the surface energy from the total energy of nucleus and the radius of tension,  $R_s$ , to determine the capillary pressure. Second, to obtain the nonzero value of Tolman length, and, consequently, the value of the curvature correction  $\Delta\sigma_{\text{curv}} \neq 0$  for a curved surface, the droplet must have the diffuse surface layer.

The value of Tolman's length could be positive or negative. Positive value of Tolman's length  $\xi > 0$  means  $\sigma_e < \sigma_\infty$  [see Eq. (1)] and negative one leads to  $\sigma_e > \sigma_\infty$  for a curved surface.

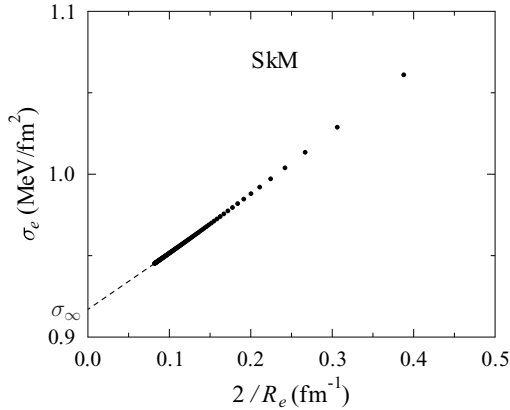


FIG. 4. Surface tension of the droplet versus the surface curvature for the range of particle number  $A = 10^2$ – $10^4$ . The calculation was performed using the SkM force.

#### IV. NUMERICAL RESULTS

Since we consider a noncharged droplet (without Coulomb), the calculation is possible up to very high values of particle number  $A \sim 10^6$ . Figure 4 shows the result of calculation for the tension  $\sigma_e$  as a function of doubled droplet curvature  $2/R_e$ . The calculation was carried out using the SkM force. Figure 4 demonstrates the negative value of  $\xi$  for this calculation. An extrapolation of curve in Fig. 4 to zero curvature  $2/R_e \rightarrow 0$  allows one to derive both the surface tension coefficient  $\sigma_\infty = \sigma_e(R_e \rightarrow \infty)$  in a planar geometry and the slope of curve which gives the Tolman length  $\xi$ . The result of such kind of extrapolation of  $\sigma_e(R_e)$  is shown in Fig. 4 by dashed line.

We have determined the Tolman’s length  $\xi$  and the planar surface tension  $\sigma_\infty$  for several sets of Skyrme interaction. For this purpose we have performed calculations up to particle number  $10^6$  and extrapolate them to zero curvature. Results are summarized in Table I. To obtain the error of the extrapolation  $2/R_e \rightarrow 0$  we estimated the magnitude of the higher order term  $\sim R_e^{-2}$  in Eq. (1). For the interval of particle numbers  $A = 10^4$ – $10^6$  we gain the term of about  $0.5R_e^{-2}$  for the SkM interaction, so one has about  $10^{-2}\%$  contribution from this term to the surface tension within the considered mass number interval. One should expect the same accuracy for the extracted values of  $\sigma_\infty$  and  $\xi$ .

We can see from Table I that Tolman’s length  $\xi$  is negative for a nuclear Fermi-liquid drop. This conclusion is also supported by the results of Ref. [3]. The value of the Tolman length is only slightly sensitive to the Skyrme force parametrization

TABLE I. Values of Tolman’s length  $\xi$  and planar surface tension  $\sigma_\infty$  obtained for different parametrizations of Skyrme forces.

Force	$\xi$ (fm)	$\sigma_\infty$ (MeV/fm <sup>2</sup> )
SkM	–0.36	0.92
SIII	–0.26	0.93
SLy230b	–0.37	1.01
T6	–0.36	1.02

with the exception of old one SIII. The calculation of the curvature correction to the surface tension by expansion around the plane surface (semi-infinite nuclear matter) was introduced in Ref. [3] and widely used for different types of nucleon-nucleon interactions with the Skyrme-type interactions among them (see, for example, [13,21].) Comparing the values of the surface,  $a_2$ , and curvature,  $a_3$ , coefficients obtained for T6 and SIII forces in Ref. [21] with the results for the same forces of Table I by means of Eqs. (35) and (37) (see the next section), we find the numerical coincidence of our results with that of Ref. [21]. The main reason of this coincidence, in our opinion, is the following. For the Gibbs-Tolman approach at the limit  $A \rightarrow \infty$  one has  $\rho_V = \rho_\infty$  and, consequently, the equimolar radius given by Eq. (18) and obtained from the condition  $\rho_S = 0$  becomes equal to the equivalent “sharp” radius of the approach proposed in Ref. [3]. In other words, the applicability of the Myers-Swiatecky approach is motivated in this case. The more detailed comparison is quite difficult since the calculation made within Gibbs-Tolman approach does not rely on the bulk asymptotics of the energy density functional.

In fact, the above comparison shows the equivalence of two approaches at large masses. It is interesting to analyze the applicability both of them for the case of small mass numbers. Following [13,21] the coefficients of mass formula are calculated using the leptodermous approximation which requires the surface layer thickness to be small as compared to the nuclear size given by the corresponding sharp radius. According to Gibbs [9] the thermodynamical relation (2) remains exact up to the zero value of  $R_s$ , provided the pressure is calculated for the matter at the value of chemical potential of the actual drop. Another conclusion concerning Gibbs-Tolman approach was made in Ref. [8], namely, the lowest limit of  $R_s$  where the definition of the surface tension make sense is about of  $R_s \sim |\xi|$ . Since both the tension and the equimolar dividing surfaces are located within the surface diffuse layer, the absolute value of  $\eta = R_e - R_s$  should not exceed the thickness,  $a$ , of the surface layer. For large masses where  $a/R \ll 1$  one can reasonably assume  $\eta$  to be approximately constant and equal to its planar limit value  $\xi$  [2,22]. For the case of very low masses of 10–20 nucleons, the approximation  $\eta \sim \xi$  as well as the leptodermous condition  $a/R \ll 1$  are violated. The Gibbs-Tolman procedure described in Sec. II defines surface quantities as the excess ones with respect to a certain volume of uniformly distributed matter which conserves the saturation property. This procedure itself does not bring any extra approximation like the leptodermous one  $a/R \ll 1$ , so the question on the applicability of the GT procedure in the case of low masses should be addressed to the model which is used for the calculation of the free energy and the chemical potential. The free energy per particle for the nuclear matter given by Eq. (15) can, obviously, be used independently on the value of mass number. One can obtain the binding energies for the range of low masses solving numerically the Euler-Lagrange equation (12) within the extended Thomas-Fermi theory [13] avoiding the use of the leptodermous condition.

Figure 5 shows the surface tension calculated within the Gibbs-Tolman procedure based on the binding energy and the chemical potential obtained from different models. In this

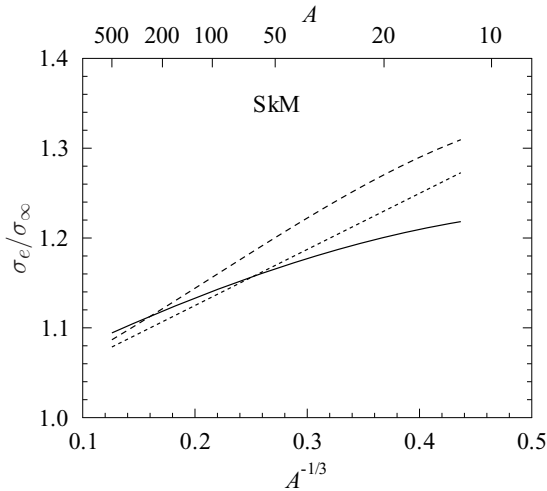


FIG. 5. Mass number dependence of the surface tension calculated using Gibbs-Tolman procedure. Solid line corresponds to the exact solution of the Euler-Lagrange equation (12) within the extended Thomas-Fermi approximation. Dashed lines show results obtained using the leptodermous expansion. Long dashed line corresponds to direct variational method with the free energy given by Eqs. (13) and (14), short dashed line is the droplet model result, see Eq. (36).

figure, we compare the result based on the exact numerical solution of the Euler-Lagrange equation (12) (solid line) with that obtained from models which use the leptodermous expansion (dashed lines). One can see from Fig. 5 that the use of  $A^{-1/3}$  expansion overestimates the value of the surface tension for nuclei with low masses. Figure 5 demonstrates that  $\sigma_e/\sigma_\infty > 1$  for finite  $A$  and the difference in predictions for presented models decreases as the mass number increases.

The effect of finite size of the drop on the surface tension value can essentially affect the yield of fragments in the nuclear multifragmentation observed for heavy ion collisions. The yield  $Y(A)$  of the fragment having a certain mass number  $A$  is given by  $Y(A) \propto \exp(-w/T)$  [10], where  $T$  is the temperature,  $w$  is the work which is needed to form the fragment from the nucleon vapor. The value of  $w$  is given by Refs. [9,10,22]

$$w = -P\mathcal{V} + \sigma\mathcal{S}. \quad (22)$$

Here, the value of the vapor pressure  $P_{\text{vap}}$  is considered to be negligible as compared to the pressure  $P$  of the liquid phase. Such simplification is possible for small drops far from the critical state. Leaving apart the temperature dependence of  $\sigma$ , let us estimate the effect of the drop size on the value of  $w$ . Using Eq. (9) one can rewrite Eq. (22) as

$$w = \frac{4\pi\sigma[R]R^2}{3} \left( 1 - \frac{\partial \ln \sigma[R]}{\partial \ln R} \right). \quad (23)$$

Using  $R_e$  for the radius of dividing surface one obtains

$$w = \frac{4\pi\sigma_e R_e^2}{3} \left( 1 - \frac{\partial \ln \sigma_e}{\partial \ln R_e} \right). \quad (24)$$

When obtaining Eq. (24) from Eq. (23), the properties of the equimolar surface were used, namely,  $\sigma'[R_e] = \sigma'(R_e)$ , see also [8]. The value of work  $w$  estimated from

TABLE II. The work of the drop formation  $w$  in units of the semi-infinite matter estimate  $w_\infty$  for mass numbers from 12 to 24, see the text for details of the calculations.  $w_{\text{DM}}$  stands for the droplet model result, see Eq. (38).

$A$	$w/w_\infty$	$w_{\text{DM}}/w_\infty$
12	1.17	1.42
16	1.19	1.39
20	1.19	1.36
24	1.20	1.34

the semi-infinite matter (no size effect) is equal to  $w_\infty = \sigma_\infty(4\pi/3)^{1/3}(A/\rho_\infty)^{2/3}$ . The comparison of  $w$  with  $w_\infty$  should apparently allocate the effect of the finite drop size. We have performed the calculation for the work of drop formation for several small mass numbers using the SkM force. The values of the free energy and the chemical potential were obtained by solving numerically the Euler-Lagrange equation (12) within the extended Thomas-Fermi theory [13,21]. Then the Gibbs-Tolman procedure was applied to evaluate  $w$  using Eq. (24). The results are presented in the second column of Table II and show values of about 20% higher than  $w_\infty$  for mass numbers from 12 to 24. This should bring the hindrance of the yield of fragments with those masses due to the effect of the finite fragment size. From the third column of Table II one can see a significant overestimation of the work  $w$  if one uses the leptodermous expansion around the value of  $w_\infty$  for the semi-infinite matter.

## V. LINK TO THE DROPLET MODEL

The Gibbs concept of dividing surface does not imply any specific energy density functional and relies on the value of the binding energy and the chemical potential which are measurable quantities. It is possible to apply this concept to the phenomenological droplet model as well. Considering the non-charged ( $N = Z$ , without Coulomb interaction) droplet at zero temperature, we will apply the same procedure as described in previous sections to extract the value of Tolman length. According to Ref. [3], one can write the free energy,  $F$ , and the chemical potential,  $\lambda$ , of the nucleus having mass number  $A$  as

$$F = -a_1 A + a_2 A^{2/3} + \left( a_3 - \frac{2a_2^2}{K} \right) A^{1/3}, \quad (25)$$

$$\lambda = -a_1 + \frac{2}{3} a_2 A^{-1/3} + \frac{1}{3} \left( a_3 - \frac{2a_2^2}{K} \right) A^{-2/3}, \quad (26)$$

where  $a_1$ ,  $a_2$ , and  $a_3$  are, respectively, the volume, the surface and the curvature correction coefficients,  $K$  is the incompressibility. From Eqs. (25) and (26) one obtains the grand potential per particle  $\omega = F/A - \lambda$  as

$$\omega = \frac{1}{3} a_2 A^{-1/3} + \frac{2}{3} \left( a_3 - \frac{2a_2^2}{K} \right) A^{-2/3}. \quad (27)$$

In terms of the droplet model, the equation of state for infinite nuclear matter beyond the equilibrium point reads

$$F/A = -a_1 + \frac{1}{2} K \epsilon^2 \quad (28)$$

and its chemical potential,  $l$ , is given by

$$l = -a_1 + \frac{1}{6}K\epsilon(9\epsilon - 2). \quad (29)$$

In Eqs. (28) and (29), the dimensionless variable

$$\epsilon = -\frac{1}{3} \frac{\rho - \rho_\infty}{\rho_\infty} \quad (30)$$

was introduced as the measure of difference between the nuclear matter density  $\rho$  and its equilibrium value  $\rho_\infty$ . Fixing the value of particle density  $\rho_V = \rho(\lambda)$  from the condition  $l(\rho) = \lambda$ , one obtains the volume part of the grand potential per particle

$$\omega_V = -\frac{2}{3}a_2A^{-1/3} - \frac{1}{3} \left( a_3 - \frac{8a_2^2}{K} \right) A^{-2/3} + \mathcal{O}(A^{-1}), \quad (31)$$

and, using also Eq. (27), the ratio  $\omega/\omega_V$

$$\frac{\omega}{\omega_V} = -\frac{1}{2} - \frac{3}{4} \frac{a_3}{a_2} A^{-1/3} + \mathcal{O}(A^{-2/3}). \quad (32)$$

Then, using Eqs. (18) and (21), one derives both radii

$$R_e = r_0A^{1/3} \left[ 1 - \frac{2a_2}{K} A^{-1/3} + \mathcal{O}(A^{-2/3}) \right], \quad (33)$$

$$R_s = r_0A^{1/3} \left[ 1 + \left( \frac{a_3}{2a_2} - \frac{2a_2}{K} \right) A^{-1/3} + \mathcal{O}(A^{-2/3}) \right] \quad (34)$$

and the Tolman length

$$\xi = -\frac{a_3}{2a_2} r_0, \quad (35)$$

where  $r_0 = (4\pi\rho_\infty/3)^{-1/3}$ . Taking the Eq. (6) at  $R = R_e$ , by the use of the Eqs. (27), (31), and (33), the surface tension reads

$$\sigma_e = \frac{(\omega - \omega_V)A}{4\pi R_e^2} = \frac{1}{4\pi r_0^2} (a_2 + a_3A^{-1/3} + \mathcal{O}(A^{-2/3})). \quad (36)$$

With the Tolman length given by Eq. (35) and the relation

$$\sigma_\infty = \frac{a_2}{4\pi r_0^2} \quad (37)$$

one can reduce Eq. (1) to Eq. (36). As seen from the above Eqs. (33), (34), and (35), both the equimolar,  $R_e$ , and the tension,  $R_s$ , radii include the term of compression effect  $(2a_2/K)A^0$ , whereas the value of Tolman's length  $\xi$  of Eq. (21) reflects purely the effect of curvature of dividing surface. Using the results presented in Table I, one may estimate the ratio of the curvature correction to the surface coefficient of the droplet model as  $a_3/a_2 \approx 0.63$  for the case of the SkM nucleon-nucleon interaction. This value of the ratio  $a_3/a_2$  is consistent with that of Ref. [4].

We have also estimated the droplet model (DM) value  $w_{\text{DM}}$  for the work needed to form the drop with mass number  $A$

from the nucleon vapor as given by Eq. (24). Using Eqs. (33) and (36) one obtains from Eq. (24) the following expression:

$$w_{\text{DM}} = \frac{a_2A^{2/3}}{3} \left[ 1 + \left( \frac{2a_3}{a_2} - \frac{4a_2}{K} \right) A^{-1/3} + \mathcal{O}(A^{-2/3}) \right]. \quad (38)$$

## VI. CONCLUSIONS

Considering a small droplet with a finite diffuse layer, we have introduced a formal dividing surface of radius  $R$  which splits the droplet onto volume and surface parts. The corresponding splitting was also done for the free energy. Assuming that the dividing surface is located close to the interface, we are then able to derive the pressure  $P$  and the surface free energy  $F_S$ . In general, the surface free energy  $F_S$  includes the contributions from the surface tension  $\sigma$  and from the binding energy of  $A_S$  particles within the surface layer. The equimolar surface and the actual physical size of the droplet was derived by the condition  $\rho_S = 0$ .

In a small nucleus, the diffuse layer and the curved interface affect the surface properties significantly. In agreement with Gibbs-Tolman concept [2,9], two different radii have to be introduced in this case. The first radius,  $R_s$ , is the surface tension radius which provides the minimum of the surface tension coefficient  $\sigma$  and the fulfillment of the Laplace relation (17) for capillary pressure. The another one,  $R_e$ , is the equimolar radius which corresponds to the equimolar dividing surface and defines the physical size of the sharp surface droplet, i.e., the surface at which the surface tension is applied. The difference of two radii  $R_e - R_s$  derives the Tolman length  $\xi$  in an asymptotic limit of large system  $A \rightarrow \infty$ . That means the presence of curved surface is not sufficient for the calculation of the curvature correction to the surface tension. The finite diffuse layer in the particle distribution is also required. We point out that the Gibbs-Tolman theory allows to treat a liquid drop within thermodynamics with minimum assumptions. Once the binding energy and chemical potential of the nucleus are known its equimolar radius, radius of tension and surface energy can be evaluated using the equation of state for the infinite nuclear matter. In this sense, in contrast to the "geometrical" definition of nuclear size [7], the Gibbs-Tolman approach does not rely on details of the particle density profile. In particular, the quantum oscillations of bulk density [11] do not need to be smoothed to obtain the volume density  $\rho_V$  which is different, in general, than the bulk density.

The sign and the magnitude of the Tolman length  $\xi$  depend on the interparticle interaction. We have shown that the Tolman length is negative for a nuclear Fermi-liquid drop. As a consequence, the curvature correction to the surface tension could lead to the hindrance of the yield of light fragments at the nuclear multifragmentation in heavy ion collisions.

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